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AN EVALUATION OF THE RESIDENCE DURATION OF MEGACRYSTS IN ALKALINE MAGMA CHAMBERS

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ABSTRACT

The integration of the differential equation of the second law of Fick applied to the diffusion of chemical elements in a semi-infinite solid made it easier to estimate the time of stay of olivine megacrysts horted into alkaline lava. The results of this research show the existence of two groups of olivine. The first remained in contact with the magmatic liquid during 30 to 34 days, while the second remained so during only 4 to 7 days only. This distinction is correlative to that based on the qualitative observation.

Key words: *Atomic diffusion, residence time, olivine megacrysts, alkaline lavas, Anti-Atlas Morocco, Canary Islands.*

RESUMEN

La integración de la ecuación diferencial de la segunda ley de Fick aplicada a la difusión en un sólido semiinfinito, permitió estimar el tiempo de la residencia de los megacristales de olivino incluidos en lavas alcalinas. Los resultados muestran que existen dos grupos de olivino: el primero persiste entre 30 a 34 días, en contacto con el líquido magmático, mientras que, el segundo, solamente lo hace entre 4 a 7 días. Esta distinción está de acuerdo con la observación petrográfica.

Palabras clave: *Difusión atómica, tiempo de residencia, megacristales de olivino, lavas alcalinas, Islas Canarias, Anti-Atlas marroquí.*

Introduction

Megacryst-sized olivine crystals are common in alkaline lavas (Irving and Frey, 1984; Liotard *et al.*, 1988). Olivine megacrysts studied here are from Saghro mountain (Anti-Atlas, Morocco) (Ibhi, 1997) and from La Palma alkaline lavas (Canary Islands) (Dautria and Girod, 1987). The origin of the olivine crystals is still debatable; however, most authors have divided them into two groups, according to their genetic relationship with the host lava (Irving, 1984; Long *et al.*, 1993):

1. Most olivine megacrysts appear in approximate from equilibrium with the host lava (Ol_E).
2. Others are in disequilibrium with the host lava (Ol_D).

Different cationic exchange models between megacrysts and host lava give the diffusion profiles upon which Fick's law is applied. The latter enables to calculate minerals period of stay in magmatic liquid.

Material and chemical analyses

Megacrysts are defined here as discrete crystals that have a dimension greater than 2 cm. They have subspherical shape or are angular fragments. The destabilisation of olivine in the host lava, produces coronitic pseudomorphs. These structures are narrow.

Megacrysts were analysed by automated microprobe, Camebax Sx 50 of CAMPARIS, by using routine analytical conditions: 15 kV acceleration voltage, 20 nA beam current, 30 s count time, and tracor Northern ZAF matrix correction with natural mineral standards. Analytical errors are ± 0.5 %-2 % for major elements and 10 %-20 % for minor elements.

Representative electron microprobe analyses of olivine megacrysts (Ol_D, Ol_E) are given in table 1; and host rock chemical analyses are given in table 2.

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Table 1.—**Representative electron microprobe analyses of olivine megacrysts (Ol_D, Ol_E)**

	Ol_{D-1}		Ol_{D-2}		Ol_{E-1}		Ol_{E-2}	
	center	rim	center	rim	center	rim	center	rim
SiO_2	40.10	37.76	40.22	38.24	39.36	38.68	38.58	37.83
FeO	13.70	24.82	13.99	26.74	19.29	24.90	20.51	24.14
MgO	45.80	37.07	45.83	34.61	41.52	36.86	41.23	36.98
CaO	0.25	0.28	0.17	0.39	0.25	0.27	0.18	0.33
MnO	0.14	0.42	0.16	0.67	0.31	0.20	0.33	0.43
NiO	0.19	0.07	0.25	0.05	0.09	0.03	0.11	0.02
Cr_2O_3	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
Total	100.10	100.40	100.60	100.70	100.50	100.90	100.80	99.70
Si	1.000	1.002	0.999	1.010	0.995	1.007	0.987	0.984
Fe	0.290	0.510	0.291	0.590	0.418	0.553	0.439	0.539
Mg	1.702	1.452	1.697	1.360	1.572	1.430	1.572	1.473
Ca	0.007	0.008	0.005	0.011	0.007	0.008	0.005	0.009
Mn	0.003	0.009	0.003	0.015	0.007	0.006	0.007	0.010
Ni	0.004	0.002	0.005	0.001	0.002	0.001	0.002	0.000
Cr	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Total	3.000	3.000	3.000	2.999	3.000	3.005	3.010	3.015
C_{Fe}	14.4	27.7	14.6	30.7	20.0	27.7	22.3	27.2

Structural formulae calculated on the basis of 4 oxygens (C_{Fe} is the mole % fayalite content of the olivine).

Diffusion equations

The analysis of the olivine megacrysts using electronic microprobe, revealed diffusion profiles of Fe and Mg (diagrams a1, b1, c1 and d1 of figures 1, 2, 3 and 4). It is known that diffusion depends on crystallographic direction, oxygen pressure, chemical composition, pression and temperature (Buerning and Buseck, 1973; Jurewicz and Watson, 1988; Nakamura and Schmalzried, 1984; Morioka and Nagazawa, 1991; Jaoul *et al.*, 1995, and Chakraborty, 1997).

Cristallographic control

Olivine is orthorhombic ($a \neq b \neq c$) and diffusion is anisotropic ($D_{[100]} \neq D_{[010]} \neq 1\ D_{[001]}$). At 1100° C, the interdiffusion coefficient ($D_{Fe\text{-}Mg}$) in olivine is 10 times greater for the c-axis than for the a and b axes (Buening and Buseck, 1973; Misener, 1974; Jurewicz and Watson, 1988).

Oxygen pressure

Dislocations and plasticity of natural olivine have been extensively investigated essentially by means of creep test experiments (Jaoul *et al.*, 1979; Gueguen and Darot, 1982, and Poumellec and Jaoul, 1984). They depend on oxygen pressure and water pressure (Poumellec and Jaoul, 1984). These mechanical properties are controlled by the diffusion rate of the slowest species D_{Si}, but with an important enhancement due to the presence of the most abundant and mobile defects, the octahedral vacancies, V_{Me}, with concentration $[V_{Me}]$, which are «pulling» the slow silicons (Jaoul, 1990).

In olivine, iron is found in considerable concentration in the two valency states (Fe^{2+} and Fe^{3+}). The vue concentration is related to oxygen fugacity (Morioka and Nagazawa, 1991) by:

$$1/2\,O_{2(gas)} + 3Fe^{x}_{Me} \Leftrightarrow 2Fe^{\bullet}_{Me} + V''_{Me} + FeO_{(solid\,solution)} \quad [1]$$

$$K = a_{FeO}\frac{(V''_{Me})(Fe^{\bullet}_{Me})^2}{{}_pO_2^{1/2}(Fe^{x}_{Me})^2} \quad [2]$$

where K is the equilibrium constant at T, pO_2 is the oxygen partial pressure around the sample and a_{FeO} represents FeO activity. Fe^{x}_{Me} is atomic fraction of iron in octahedral sits, $Fe^{\bullet}_{Me}$ stands for the Fe^{3+} concentration in Fe^{2+} site and V''_{Me} represents vacant octahedral sits.

Electronic neutrality $2(V''_{Me}) = (Fe^{\bullet}_{Me})$ yields:

$$D_{Fe/Mg}a(V''_{Me})a_PO_2^{1/6} \quad [3]$$

The Fe/Mg interdiffusion coefficient ($D_{Fe/Mg}$) depends on pO_2 roughly as $(pO_2)^{1/6}$. The relationship has been verified experimentally by Buening and Buseck (1973) and Nakamura and Schmalzried (1983) (the latter found a 1/5.5 exponent in their study).

Table 2.—**Host-rock chemical analyses [1: % oxide weight; 2: % atom weight = (% oxide weight × atomic weight)/molecular weight].**

	Host-rock (Ol_{D-1}, Ol_{D-2} and Ol_{E-2})				Host-rock (Ol_{E-1})		
	(1)		(2)		(1)		(2)
SiO_2	40.43	Si	18.896	SiO_2	38.41	Si	17.952
FeO	10.98	Fe	8.534	FeO	11.53	Fe	8.962
MgO	10.73	Mg	6.471	MgO	12.85	Mg	7.749
CaO	15.35	Ca	10.969	CaO	12.27	Ca	8.768
MnO	0.20	Mn	0.154	MnO	0.17	Mn	0.131
Al_2O_3	11.76	Al	6.223	Al_2O_3	12.96	Al	6.858
TiO_2	2.78	Ti	1.666	TiO_2	3.08	Ti	1.846
Na_2O	3.36	Na	2.492	Na_2O	3.23	Na	2.396
K_2O	1.80	K	1.494	K_2O	1.04	K	0.863
P_2O_5	1.07	P	0.467	P_2O_5	0.72	P	0.314
PF	2.19			PF	1.32		
Total	100.60			Total	99.58		

Host lava oxygen fugacity has been determined from ilmenite-magnetite couple (temperature/oxygen fugacity curves; Buddington and Lindsley, 1964). At 1,100° C ± 40° C, this fugacity is of $10^{-10.4 \pm 0.6}$ atm.

Diffusion coefficient

Following Nakamura and Schmalzried (1983), we have considered that D_{Fe-Mg} varies exponentially with X_{Fe} so that we have

$$D^{Ol}_{Fe-Mg} = D^{Fo}_{Fe-Mg} \exp(\in X_{Fe}) \quad [4]$$

where X_{Fe} is the Fe/(Fe + Mg) content of the olivine, D^{Fo}_{Fe-Mg} is the Fe-Mg interdiffusion coefficient in an olivine of forsterite (symbol Fo) composition, and $\in$ is a factor empirically evaluated. Nakamura and Schmalzried have verified this relationship at higher temperatures (1,050° <T <1,280° C) by treating their data with the Boltzmann-Matano approach. For each experimental temperature they determined a value of $\in$, with $\in$ increasing from 4.14 at 1,050° C to 6.38 at 1,280° C.

The expression for D_{Fe-Mg} determined by Buening and Buseck (1973) at a temperature below 1125° C is given by the expression:

$$D_{Fe-Mg} = 10^2 (fO_2)^{1/6} \exp(-0.0501 C_{Fe} - 14.03) \exp[(-31.66 + 0.2191 C_{Fe})/RT] \quad [5]$$

where D_{Fe-Mg} is in units $cm^2\ s^{-1}$, C_{Fe} is the mole % fayalite content of the olivine, R is the gas constant and T is the absolute temperature. This is the expression that we used for our calculations.

Temperature - Pression estimates

In alkaline basalt, the olivine appears at liquidus. The host lava temperature is calculated by using Leeman and Schridegger thermometric equations (1977). This was done by supposing that the equilibrium is realised between the rock and the most magnesic olivine phenocrysts.

Regression formulas are the following:

$$\ln D_{Fe2+} = 9016/T - 5.46 \text{ and } \ln D_{Mg} = = 8916/T - 4.29 \quad [6]$$

Obtained temperatures are 1,100° C ± 40° C.

Pression estimation have been indirectly performed from previously calculated temperatures, by using the geotherm established by Mercier and Carter (1975). They are 2.9 GPa ± 0.1 GPa.

Numerical application

The crystal surface is supposed plane. That means its irregularities are small compared to the depth of the concentration gradient. The diffusion coefficient is supposed constant in all points of the system, because the variations of the temperature, of oxygen fugacity and those of chemical compositions are slow in the magmatic chamber.

The solution to the diffusion problem is the following, according to the boundary conditions:

$$C_{(x,t)} = C_1 + (C_0 - C_1)\,\mathrm{erf}\left(\frac{x}{2\sqrt{Dt}}\right) = = C_1 + (C_0 - C_1)\,\mathrm{erf}(ax) \quad [7]$$

$$\text{with } a = \left(\frac{1}{2\sqrt{Dt}}\right)$$

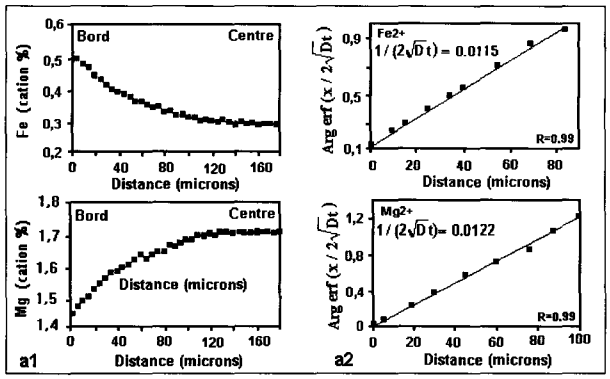

Fig. 1.—a1: Fe and Mg diffusion profiles in olivine megacrysts Ol_{D-1}. a2: Concentration data according to distance, after error function inversion.

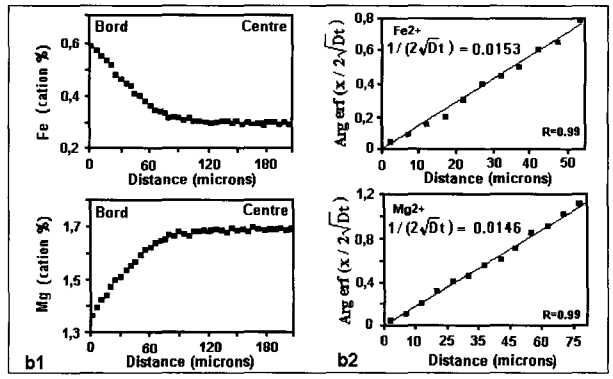

Fig. 2.—b1: Fe and Mg diffusion profiles in olivine megacrysts Ol_{D-2}. b2: Concentration data according to distance, after error function inversion.

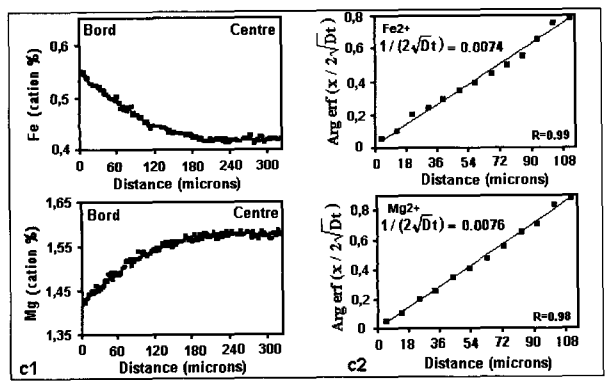

Fig. 3.—c1: Fe and Mg diffusion profiles in olivine megacrysts Ol_{E-1}. c2: Concentration data according to distance, after error function inversion.

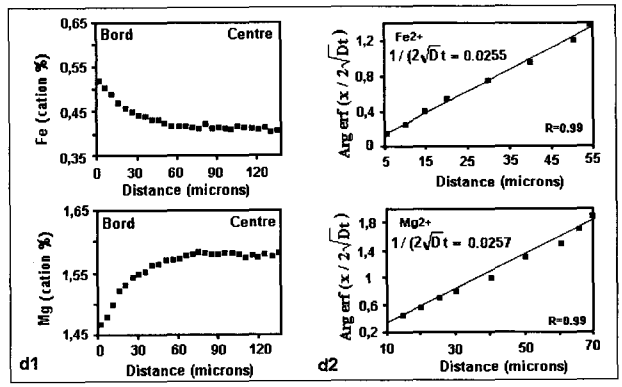

Fig. 4.—d1: Fe and Mg diffusion profiles in olivine megacrysts Ol_{E-2}. d2: Concentration data according to distance, after error function inversion.

In equation [7], C_1 is olivine superficial concentration and C_0 the initial concentration in the crystal (the concentration at the crystal core). x is the diffusion distance, D is the diffusion coefficient, and t diffusion duration; erf is the error function.

[7] is rewritten as

$$\mathrm{erf}^{-1}\left[\frac{C_{(x,t)}-C_1}{C_0-C_1}\right]=ax \qquad [8]$$

in which "a" is the slope of the line.

The argument of the erf function must have a linear relationship with x in classical diffusion. If not, there is some problem across the interface for Fe/Mg exchange.

Result and conclusion

The studied megacrysts show that the arg erf $\left(\frac{x}{2\sqrt{Dt}}\right)$ in relation to the diffusion distance have correlation coefficients around 0.99 (diagrams a2, b2, c2 and d2 of figures 1, 2, 3 and 4).

The application of the Fick's law on cationic diffusion profiles made it possible for us to calculate the time spent by these crystals in the lava. The different olivine crystals residence duration is calculated and reported in table 3. The results show the existence of two olivine populations. The first was in contact with the magmatic liquid from 30 to 34 days for olivine in equilibrium with host lava (Ol_E), while the second was only for 4 to 7 days, regrouping crystals in disequilibrium (Ol_D).

Megacrysts in desiquilibrium remained in contact with host lava for just the period reminding that of cropping up of a xenolith having a diameter of 10 cm in volcanic chimney calculated from Stockes law, for a 50 km distance (Kushiro *et al.*, 1976). The origin of these megacrysts can be looked for in the desegregation of Cr-diopside suite xenoliths. This goes side by side with Johnson's chemical studies (1989).

Nakamura (1995) estimated the residence time of olivine phenocrysts in magma that erupted from the

Table 3.—**Olivine megacrysts period of stay calculation in alkaline lavas (T = 1,100° C).**

Group A	Axis	C_{Fe}	$D(\mu m^2/s)$	$\frac{1}{2\sqrt{Dt}}$	t (hours)
Ol_{D-1}					
Mg	[001]	14.4 ± 0.2	$3.27\ 10^{-3}$ [18]*	0.0122 ± 0.003	142 ± 84
Fe	[001]	14.4 ± 0.3	$3.27\ 10^{-3}$ [20]*	0.0115 ± 0.004	160 ± 127
Ol_{D-2}					
Mg	[001]	14.6 ± 0.2	$3.18\ 10^{-3}$ [18]*	0.0146 ± 0.003	102 ± 49
Fe	[001]	14.6 ± 0.3	$3.18\ 10^{-3}$ [20]*	0.0153 ± 0.002	93 ± 31

Group B	Axis	C_{Fe}	$D(\mu m^2/s)$	$\frac{1}{2\sqrt{Dt}}$	t (hours)
Ol_{E-1}					
Mg	[001]	20.0 ± 0.3	$1.71\ 10^{-3}$ [20]*	0.0076 ± 0.003	700 ± 430
Fe	[001]	20.0 ± 0.4	$1.71\ 10^{-3}$ [22]*	0.0074 ± 0.003	738 ± 434
Ol_{E-2}					
Mg	[100]	22.3 ± 0.3	$1.34\ 10^{-4}$ [25]*	0.0257 ± 0.005	798 ± 440
Fe	[100]	22.3 ± 0.4	$1.34\ 10^{-4}$ [27]*	0.0255 ± 0.006	811 ± 480

D is calculated from equation [5] (* values in brackets are percent error).

northern Yatsugatake volcanoes in Japan using compositional zoning. Just from the results of modelling the Fe/Mg compositional profiles; he concluded that the residence times of olivines was between 10 days and 1 year.

We think, based on these results, that it becomes possible to talk about the notion of the period of stay of olivine megacrysts once the distinction between the form more or less in equilibrium and those in disequilibrium with host liquid is established.

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